

U.S. Patent Application Serial No. **10/530,475**
Amendment filed June 27, 2007
Reply to OA dated February 20, 2007

REMARKS

Claims 1-17 are presented for examination. Claim 4 has been cancelled. In order to more particularly point out and distinctly claim the subject matter to which the applicants regards as their invention, claim 1 has been amended to include the limitations of original claim 4.

The applicants respectfully submit that no new matter has been added. It is believed that this Amendment is fully responsive to the Office Action dated **February 20, 2007**.

Claims 1, 2, 3, and 5-17 define a porous semiconductor film forming process and use of a semiconductor film formed thereby.

As described in the present specification, page 19, line 29, to page 21, line 4, the semiconductor film forming process of the present invention achieves the following remarkable effects, in particular by applying a specific semiconductor particle dispersion liquid to a substrate by spray coating in such a manner that the atomized droplets of the dispersion liquid have a mean diameter of about 30 μm or less. Since semiconductor particles or small aggregates thereof deposited on the substrate form a semiconductor film, it is possible to form a porous film having excellent adhesion to the substrate and high strength.

When the solvent used in the semiconductor particle dispersion liquid consists only or mainly of ethanol, the dispersion liquid is stable because of the high affinity of ethanol to the

semiconductor particles. Therefore, the dispersion liquid, even when being subjected to a high shear stress at the time of spray coating, is unlikely to cohere at the nozzle tip and thereby clog the nozzle.

Further, the high affinity of ethanol facilitates necking of the coating film formed from the semiconductor particle dispersion liquid, and thus even when the dispersion liquid is applied to a flexible substrate such as a high polymer film, it does not peel off when the substrate flexes.

Since the solvent partially evaporates from the dispersion liquid during spray coating, the semiconductor particles being sprayed are stabilized.

The process is capable of forming a porous semiconductor film at low temperatures not higher than 200 °C, and therefore can be carried out using a thermoplastic resin substrate such as a high polymer film.

In particular, by microwave sintering of the semiconductor particle dispersion liquid applied to a high polymer film substrate such as a polyethylene terephthalate, a porous film can be formed in which the semiconductor particles are uniformly sintered, even when the film has a large area.

When a porous film of a semiconductor with photocatalytic activities, such as titanium oxide, is formed on a substrate by the process of the present invention, a photocatalyst or photoelectrode for dye-sensitized solar cells with excellent performance can be advantageously obtained.

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Claims 1, 4-9 and 12-14 are rejected under 35 USC §103(a) as being unpatentable over Akui et al. (U.S. Patent Application Publication 2002/0042343).

Claim 4 has been cancelled.

With respect to claim 1, the Office Action states that **Akui** discloses a semiconductor film forming process comprising the steps of applying a semiconductor particle dispersion liquid to a substrate surface by spray coating, and drying the coating to form a porous semiconductor film (see Akui, ¶ [0009], [0021], and [0047]), and that the semiconductor particle dispersion liquid used in **Akui** is a dispersion in methanol or ethanol of particles of a semiconductor, specifically a metal oxide (see Akui, ¶ [0037] and [0033]).

The applicants respectfully disagree that the claimed invention reads on **Akui**. The cited prior art discloses a coating composition for forming a titanium oxide film, the composition comprising an aqueous peroxy titanate acid solution (A) and polyethylene glycol (B) (for example, see Akui, claim 1), and describes an embodiment in which the peroxy titanate acid solution is obtained in the presence of a titanium oxide sol (see Akui, ¶ [0035]). In **Akui**, peroxy titanate acid is used in the form of an aqueous solution, not in the form of a dispersion of semiconductor particles, and **Akui** merely states that methanol, ethanol, or the like, can optionally be used for preparing the aqueous peroxy titanate acid solution (see Akui, ¶ [0033]). Further, the titanium oxide sol cannot be used singly, since the sol shows poor film-forming properties (see Akui, ¶ [0036]). The titanium oxide sol comprises titanium oxide particles dispersed in water, and use of an alcohol in the sol is only optional (see Akui, ¶ [0037]).

Furthermore, as acknowledged by the Office Action, **Akui** teaches nothing about the mean diameter of the atomized droplets during spray coating.

In contrast, in the semiconductor film forming process defined in amended claim 1 of the present application, a dispersion in methanol and/or ethanol of particles of at least one semiconductor selected from the group consisting of metal oxides, perovskites, metal sulfides and metal chalcogenides, is applied by spray coating in such a manner that the atomized droplets of the dispersion have a mean diameter of 30 μm or less, and dried to form a porous semiconductor film.

Such a process of the present invention achieves the above-mentioned remarkable effects, namely the following:

- 1 . Since semiconductor particles or small aggregates thereof deposited on the substrate form a semiconductor film, it is possible to form a porous film having excellent adhesion to the substrate and high strength
- 2 . When the solvent used in the semiconductor particle dispersion liquid consists only or mainly of ethanol, the dispersion liquid is stable because of the high affinity of ethanol to the semiconductor particles. Therefore, the dispersion liquid, even when being subjected to a high shear stress at the time of spray coating, is unlikely to cohere at the nozzle tip and thereby clog the nozzle.

Further, the high affinity of ethanol facilitates necking of the coating film formed from the semiconductor particle dispersion liquid, and thus even when the dispersion liquid is applied to a flexible substrate such as a high polymer film, it does not peel off when the substrate flexes.

- 3 . Since the solvent partially evaporates from the dispersion liquid during spray coating, the semiconductor particles being sprayed are stabilized.
- 4 . The process is capable of forming a porous semiconductor film at low temperatures not higher than 200 $^{\circ}\text{C}$, and therefore can be carried out using a thermoplastic resin substrate such as a high polymer film.

In particular, the effect (1), i.e., formation of a porous semiconductor film having excellent adhesion to the substrate and high strength, can be achieved by using volatile methanol and/or ethanol as dispersion media and spraying the dispersion liquid in such a manner that the atomized droplets have a small mean diameter of 30 μm or less, because, under these conditions, the semiconductor particles deposited on the substrate surface have an increased binding energy due to the sharp decrease of their kinetic energy. Examples 1 to 3 and Comparative Example 2 in the present specification demonstrate such unexpected effects achieved by the small mean diameter of the atomized droplets.

The process of the present invention also has the above effect (4), i.e., the process is capable of forming a porous semiconductor film at low temperatures not higher than 200 $^{\circ}\text{C}$, and therefore can be carried out using a thermoplastic resin substrate such as a high polymer film. In contrast, in **Akui**, the applied coating composition is calcined at a temperature of not lower than 200 $^{\circ}\text{C}$ (Akui, ¶ [0038]) so that the peroxotitanic acid is converted into titanium oxide and numerous pores are produced upon volatilization of polyethylene glycol, rendering the titanium oxide film porous (Akui, ¶ [0043]). This means that it is impossible to obtain a porous film at a low temperature not higher than 200 $^{\circ}\text{C}$. Therefore, the effect (4) of the present invention cannot be expected from **Akui**.

Akui fails to teach or suggest the claimed invention because the reference does not disclose a process comprising applying a specific semiconductor particle dispersion liquid by

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spray coating in such a manner that the atomized droplets have a mean diameter of 30 μm or less and drying the coating to form a porous semiconductor film, as defined in claim 1, or the remarkable effects (1-4) discussed above.

In light of the foregoing, the applicants respectfully request that this rejection be reconsidered and withdrawn.

Claims 2 and 3 are rejected under 35 USC 103(a) as being unpatentable over Akui, and further in view of Arakawa et al. (U.S. Patent No. 6,228,796).

Arakawa is cited for the disclosure of an organic-inorganic hybrid material comprising a substrate (e.g., a thermoplastic resin), an intermediate layer, and a photo-catalytic layer.

Arakawa merely discloses an organic-inorganic hybrid material comprising an intermediate layer and a photo-catalysis layer formed on a high polymer film. Therefore, even those skilled in the art would not have easily conceived of the process defined in claims 2 and 3 of the present application by combining **Akui** with **Arakawa**. Furthermore, **Arakawa** fails to overcome the deficiencies of **Akui** discussed above.

Given that there is no motivation for one having ordinary skill in the art to combine **Akui** and **Arakawa**, and in light of the foregoing discussion of **Akui**, the applicants respectfully request that this rejection be reconsidered and withdrawn.

Claims 10 and 11 are rejected under 35 U.S.C. § 103(a) as being unpatentable over

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Akui and further in view of Kawazu, et al. (U.S. Patent Application Publication 2002/0186469).

Kawazu is cited for the disclosure of drying a coating with irradiation with electromagnetic waves.

Kawazu merely discloses formation of a fine metal particle dispersion film by applying a sol coating liquid to a transparent substrate and irradiating the applied sol coating liquid with electromagnetic waves. Therefore, even those skilled in the art would not have easily conceived of the process defined in claims 10 and 11 of the present application by combining **Akui** with **Kawazu**. Furthermore, **Kawazu** fails to overcome the deficiencies of **Akui** discussed above.

Given that there is no motivation for one having ordinary skill in the art to combine **Akui** and **Kawazu**, and in light of the foregoing discussion of **Akui**, the applicants respectfully request that this rejection be reconsidered and withdrawn.

Claims 15-17 are rejected under 35 U.S.C. § 103(a) as being unpatentable over Muramatsu (U.S. Patent Application Publication 2002/0042342) and in view of Akui.

Muramatsu merely discloses a photocatalyst for photoelectrodes, which comprises a support, such as glass, and a metallic oxide layer provided on the support by applying and baking a colloidal solution including at least a metallic oxide precursor. Therefore, even those skilled in the art would not have easily conceived of the process defined in claims 15-17 of the present application by combining **Akui** with **Muramatsu**. Furthermore, **Muramatsu** fails to overcome

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the deficiencies of **Akui** discussed above.

Given that there is no motivation for one having ordinary skill in the art to combine **Akui** and **Muramatsu**, and in light of the foregoing discussion of **Akui**, the applicants respectfully request that this rejection be reconsidered and withdrawn

In view of the aforementioned amendments and accompanying remarks, claims 1, 2, 3, and 5-17, as amended, are in condition for allowance.

If, for any reason, it is felt that this application is not now in condition for allowance, the Examiner is requested to contact the applicants undersigned attorney at the telephone number indicated below to arrange for an interview to expedite the disposition of this case.

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In the event that this paper is not timely filed, the applicants respectfully petition for an appropriate extension of time. Please charge any fees for such an extension of time and any other fees which may be due with respect to this paper, to Deposit Account No. 01-2340.

Respectfully submitted,

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